

Composite Polarons in Ferromagnetic Narrow-Band Metallic Manganese Oxides

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Abstract

Moving electrons accompanied by Jahn-Teller phonon and spin-wave clouds may form composite polarons in ferromagnetic narrow-band manganites. The ground-state and the finite-temperature properties of such composite polarons are studied in the present paper. By using the variational method, it is shown that the energy of the system at zero temperature decreases with the formation of the composite polaron and the composite polaron behaves as a Jahn-Teller phononic polaron; the energy spectrum of the composite polaron at finite temperatures is found to be strongly renormalized by the temperature and the magnetic field. It is suggested that the composite polaron contributes significantly to the transport and the thermodynamic properties in ferromagnetic narrow-band metallic manganese oxides.

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I. INTRODUCTION

Because of the electron-lattice interaction, an electron moving in a polarized or a dynamically distorted lattice surrounded by phonon clouds at low temperature is damped and forms a phononic polaron [1]. Similarly, a moving electron in ferromagnetic (or antiferromagnetic) background disturbs the nearby local spin field and excites spin waves, therefore an electron surrounded by spin-wave clouds can form a magnetic polaron [2]. One could anticipate that in the presence of strong electron-lattice interaction and electron-spin coupling, a moving electron clouded both by phonons and spin waves can form a kind of new quasiparticle, which we call a composite polaron. In the presence of a magnetic field and for certain temperature, the physical properties of a system with composite polarons differ from those of systems with either magnetic polarons or the phononic polarons. Since there exist strong electron-phonon interactions, arising from the Jahn-Teller effect of MnO_6 , and strong electron-local spin interactions, arising from Hund's rule coupling, this kind of new quasiparticle might be found in ferromagnetic manganese oxide materials, such as the doped lanthanum manganites and neodymium manganites.

Recently the colossal magnetoresistance (CMR) effect has been found in manganites, such as $La_{1-x}Ca_xMnO_3$ and $Nd_{1-x}Sr_xMnO_3$ [3-6]. The huge magnetoresistance change of more than 3-6 orders in magnitude has potential technical applications. Other unusual transport, magnetic, and thermodynamic properties and the microscopic mechanism of the CMR effect have attracted great interest theoretically and experimentally. The crystal and magnetic structures of $La_{1-x}R_xMnO_3$ systems had been studied in 1950s by the x-ray crystallography and neutron diffraction technique [7,8]. In manganites, an Mn ion is surrounded by six O^{2-} ions and forms an octahedron. Due to the crystalline field effect, the 3d energy level of the Mn ion is split into a low-lying triplet (t_{2g}) and a high-energy doublet (e_g). Therefore, in $La_{1-x}R_xMnO_3$, the three d electrons of Mn^{3+} and Mn^{4+} ions fill the d- t_{2g} band (the filled band) and form a localized core spin $S=\frac{3}{2}$ via strong intraatomic Hund's rule coupling. The extra d electrons in Mn^{3+} fill the higher d- e_g band and interact with t_{2g} electrons through strong

Hund's coupling. These two bands are separated by about 1.5 eV [9]. The localized spins tend to align parallel through the double exchange interaction between Mn^{3+} and Mn^{4+} ions and form a ferromagnetic background [10, 11]. Electrons in the d- e_g band hop between Mn ions as itinerant ones and are responsible for the electric conduction in these systems. The degenerate d- e_g doublet of Mn^{3+} in MnO_6 octahedron will be further split due to the Jahn-Teller effect [12, 13], the distortion of oxygen atoms reduces the symmetry of the MnO_6 octahedron and stabilizes it. Electron moving together with the distorted crystalline field (dynamical Jahn-Teller distortion) will form a polaron. Recent experiments and theories [13-18] have confirmed the existence and the importance of the Jahn-Teller distortion in $\text{La}_{1-x}\text{R}_x\text{MnO}_3$ compounds. Millis et al. [13] suggested that the dynamic Jahn-Teller effect plays a crucial role for the physical properties of manganites. In the meantime, doped $\text{La}_{1-x}\text{R}_x\text{MnO}_3$ ($0.1 < x < 0.5$) are ferromagnetically ordered at temperatures below the Curie temperature T_c . The exchange coupling between the mobile electron and ordered localized spins can form a magnetic polaron. Therefore the motion of an electron at the e_g level in lanthanum manganites may form a composite quasiparticle, with the coexistence of phononic and magnetic polaron simultaneously in the same place, this called the composite polaron.

The composite polaron (CP) in the present situation is small in the metallic regime. An earlier study [19] had shown that the lattice deformation can stabilize small magnetic polaron, so the CP formed in ferromagnetic narrow-band metallic manganese-oxides is stable. In this paper in Sec. II and Sec. III, the framework of the composite polaron is developed for perovskite-type manganese-oxides and its dependence on the temperature and the magnetic field are discussed. We found that the formation of a CP in manganese oxide systems is favorable in energy, and propose that the CP plays an essential role for the transport and the thermodynamic properties of lanthanum manganites. The formation of the CP could be responsible for the microscopic mechanism of the CMR effect. Finally, we draw conclusions in Sec. IV.

II. Composite Polaron at Zero Temperature

We first discuss the possibility of the formation of CP in ferromagnetic narrow-band manganese-oxides at zero temperature. The typical physical processes in manganites can be decomposed into three parts: the electron-electron interaction (the hopping process and the Coulomb interaction), H_0 , the electron-lattice interaction (Jahn-Teller effect), H_{e-ph} , and the interaction between localized spins and mobile electrons (Hund's coupling), H_{e-m} . So the Hamiltonian is:

$$H = H_0 + H_{e-m} + H_{e-ph} \quad (1)$$

$$H_0 = \sum_{ia\sigma} [\epsilon_d^a - \sigma\mu_B B] d_{ia\sigma}^\dagger d_{ia\sigma} + \sum_{\langle ij \rangle ab\sigma} t^{ab} d_{ia\sigma}^\dagger d_{jb\sigma} \quad (2)$$

$$H_{e-ph} = \sqrt{\alpha\hbar\omega} \sum_{ia} n_{ia} \beta_a [b_i^\dagger + b_i] + \sum_i \hbar\omega b_i^\dagger b_i \quad (3)$$

$$H_{e-m} = -J_H \sum_{ia\mu\nu} \mathbf{S}_i \cdot d_{ia\mu}^\dagger \sigma_{\mu\nu} d_{ia\nu} - \sum_{\langle ij \rangle} A_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \sum_i g\mu_B B S_i^z \quad (4)$$

In Eq.(2), $d_{ia\sigma}^\dagger$ creates an e_g electron at site \mathbf{R}_i at level a with spin σ , t^{ab} denotes the hopping integral of the e_g electron from one site to its nearest-neighbor, ϵ_d is the site energy of the mobile electron with respect to the chemical potential, a (or b) denotes the energy-level index of Jahn-Teller splitting in e_g level. In Eq.(3), b_i^\dagger creates a Jahn-Teller phonon at site \mathbf{R}_i with mode $\hbar\omega$, α represents the electron-phonon coupling constant, β_a is a two-component constant vector, it is -1 or 1 for the low or the high level of the Jahn-Teller splittings, respectively. Several experiments have shown that in the metallic regime, the magnetic ordering of the local spins is of Heisenberg ferromagnetic type [20, 21]. In Eq.(4), A_{ij} is the effective ferromagnetic exchange constant between manganese spins with only the nearest-neighbor interaction being considered, $-g\mu_B B$ represents the Zeemann energy in magnetic field \mathbf{B} . The mobile electron is scattered from state $i\nu$ to state $i\mu$ by the localized spin \mathbf{S}_i due to the Hund's coupling J_H between the mobile electrons and the core spins. As suggested by Kubo and Ohata [22], Millis et al [13] and Zang et al [14], $J_H \gg t^{ab}$, so the bandwidth of the metallic manganites is narrow.

In doped perovskite manganites, the double exchange interaction due to the hole hopping and the strong Jahn-Teller electron-phonon coupling is limited to a range of

a few lattice constant. This fact suggests that small polaron picture is suitable for the CMR materials. Through a canonical transformation, $H' = e^{-S} H e^S$, one could eliminate the linear term in phonon degrees of freedom in Eq. (1). By choosing $S = \sum_i \sqrt{\alpha/\hbar\omega} n_{i\alpha} \beta_\alpha (b_i^\dagger - b_i)$, an effectively attractive electron-electron interaction is introduced and the Hamiltonian Eq. (1) can be rewritten as:

$$\begin{aligned} H' = & \sum_{ia\sigma} \epsilon_{i\sigma}^a d_{ia\sigma}^\dagger d_{ia\sigma} + \sum_{\langle ij \rangle > ab\sigma} t^{ab} d_{ia\sigma}^\dagger d_{jb\sigma} \hat{X}_{ia}^\dagger \hat{X}_{jb} \\ & - \sum_{ia,b} \alpha n_{ia} \beta_a n_{ib} \beta_b + \sum_i \hbar\omega_i b_i^\dagger b_i + H_{e-m} . \end{aligned} \quad (5)$$

Where $\epsilon_{i\sigma}^a = \epsilon_d^a - \mu - \sigma\mu_B B$, and $\hat{X}_{ia}^\dagger = \exp[\sqrt{\alpha/\hbar\omega} \beta_a (b_i - b_i^\dagger)]$. In the present studies we are only interested in the ground state of the CP so it is reasonable to assume that the mobile electrons stay in the lower level of Jahn-Teller splittings and we will drop the index a and b in Eq. (5). The term $-\sum_{ia,b} \alpha n_{ia} \beta_a n_{ib} \beta_b$ is reduced to a renormalization shift of the bare energy level by $-\alpha$ and the energy shift of the electrons from the electron-phonon interaction, α , is absorbed into ϵ_d^a .

We choose a set of basis function to construct the variational ground-state wavefunction of the CP at T=0 K:

$$|\psi_i(S_0^z) \rangle = \sqrt{\frac{S + S_{0i}^z + 1}{2S + 1}} [d_{i\uparrow}^\dagger + \frac{d_{i\downarrow}^\dagger S_i^\dagger}{S + S_{0i}^z + 1}] \Pi_{j=1}^N |S_j S_j^z \rangle |0 \rangle_{e-ph} \quad (6)$$

where $\Pi_{j=1}^N |S_j S_j^z \rangle$ denotes the ferromagnetic background and $|0 \rangle_{e-ph}$ represents the electron and the phonon vacuum state. S_{0i}^z denotes the mean value of z-component of the local spin at R_i . This basis set is constructed from the wavefunction of the vacuum state of phonon, $|0 \rangle_{ph}$, and that of the magnetic polaron (Ref. [2]): $|\chi \rangle = \sqrt{\frac{S + S_{0i}^z}{2S + 1}} [d_{i\downarrow}^\dagger - d_{i\uparrow}^\dagger S_i^- / (S + S_{0i}^z)] \Pi_{j=1}^N |J_j J_j^z \rangle |0 \rangle_e$. If we let $\alpha \rightarrow 0$ or $J_H \rightarrow 0$ in (5), the problem reduces to that of the magnetic polaron or the phononic polaron, respectively.

The ground state wavefunction of the CP can be constructed in terms of the basis set, $|\psi_i(S_0^z) \rangle$, through a linear combination:

$$|G \rangle = \sum_{i S_0^z} c_i(S_0^z) |\psi_i(S_0^z) \rangle \quad (7)$$

here $c_i(S_0^z)$ is the variational coefficient of the ground-state wavefunction, S_0^z represents all spin variables. Acting the Hamiltonian H' on $|G \rangle$ gives to the ground-state energy

of the CP: $E_g < G|G \rangle = \langle G|H'|G \rangle$. Minimizing the above expression with respect to the coefficients $c_i(S_0^z)$, one gets:

$$E_g c_i = -J_H S c_i + t e^{-\frac{p}{2}} \sum_{\delta} \frac{\sqrt{(S + S_{0i}^z + 1)(S + S_{0i+\delta}^z + 1)}}{2S + 1} c_{i+\delta} \quad (8)$$

$$+ \frac{S + S_{0i+\delta}^z + 1}{2S + 1} [\epsilon_{\uparrow} + \epsilon_{\downarrow} \frac{S - S_{0i}^z}{S + S_{0i+\delta}^z + 1}] c_i + \frac{S - S_{0i}^z}{2S + 1} [-g\mu_B B - 2A \sum_{\delta} S_{0i+\delta}^z] .$$

After taking Fourier transformation, the nontrivial solution of the coefficients $c_i(S_i^z)$ gives rise to the ground-state energy spectrum of the CP:

$$E_g(k) = \epsilon_d - J_H S + z t e^{-\frac{p}{2}} \gamma(k) \frac{\sqrt{(S + S^z + 1)(S + S_{\delta}^z + 1)}}{2S + 1} \quad (9)$$

$$- \frac{2S}{2S + 1} [\mu_B B + \frac{g}{2S} \mu_B B + A \sum_{\delta} S_{\delta}^z]$$

where $p = \alpha/\hbar\omega$ is the relative strength of the electron-phonon interaction, z is the partition number, and $\gamma(k)$ the structure factor. The physical meaning of each term in the above expression is obvious: the first two terms, $\epsilon_d - J_H S$, came from the intraatomic interaction, and the third term describes the renormalized energy dispersion. The electron-phonon interaction and the spin-electron coupling narrow the bare energy spectrum of the electrons, $z t \gamma(k)$, by a factor of $\exp(-p/2)$ and a factor of $\frac{\sqrt{(S+S_0^z+1)(S+S_{0\delta}^z+1)}}{2S+1}$, respectively. However one notices that at the absolute zero temperature, the system is ferromagnetically ordered and there is no spin excitation, $S^z=S$, so that $\frac{\sqrt{(S+S_0^z+1)(S+S_{0i+\delta}^z+1)}}{2S+1} = 1$. Therefore the CP behaves as phononic polaron at zero temperature. This point lies in the following fact that at zero temperature, there does not exist any spin deviation in ferromagnets so the electron is only accompanied by dynamical distorted lattice field. However the zero-temperature CP differs from the Jahn-Teller polaron in the intraatomic Hund's coupling (the second term in Eq. (9)) and the effective Zeemann energy of the CP (the last term in Eq. (9)). It can be seen that the formation of the CP lowers the energy of the system significantly, so it is more favorable in energy.

The presence of strong Jahn-Teller electron-phonon coupling in ferromagnetic perovskite manganites has been confirmed by several experiments [15-18]. By using neutron powder-diffraction data [15], it is shown that the static distortion of oxygen around

manganese resulting from Jahn-Teller electron-phonon coupling is about 0.12 \AA in $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$ ($x \approx 0.2$). The direct support of the presence of electron-phonon coupling came from the oxygen isotope experiments done by Zhao et al. [23]. It is believed that the ferromagnetic coupling between manganese spins comes from the double-exchange interaction, mediated through oxygen atoms, so the motion of oxygen atoms will affect the double-exchange strength, hence the ferromagnetic coupling, by a factor $e^{-p(1/2+\langle n_{ph} \rangle)}$ (here $\langle n_{ph} \rangle$ is the mean phonon number at finite temperatures, see next section). Since $p \sim 1/\omega \sim M^{1/2}$, M is the mass of oxygen, the heavier the oxygen nucleus is, the weaker the coupling is. Therefore, the presence of a strong electron-phonon interaction will lead to a significant decrease of the Curie temperature. Zhao et al. showed that the oxygen isotope exponent, $\alpha_o = -d\ln T_c/d\ln M_o$, is as high as 0.85 for $\text{La}_{0.8}\text{Ca}_{0.2}\text{MnO}_3$, suggesting the existence of strong electron-phonon interaction. Accordingly, one could estimate that the relative strength of electron-phonon interaction, p , is about $1 - 2$, which belongs to the strong coupling regime.

On the other hand, besides the strong electron-phonon interaction, the spin-electron interaction is also pretty strong. In the model Hamiltonian Eq.(4), the Hund's coupling between the carrier and the local spin is about 5 eV, much larger than the conduction bandwidth $2zt$ ($\approx 2.0 \text{ eV}$ [13]). Experiments done by Kuster [3], von Helmolt [4], Jin [5] and other groups proposed that spin polaron may dominate the electric transport. Such viewpoint is supported by the fact that the logarithm of the conductivity of $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$ exhibits $T^{-1/4}$ dependence on temperature [24], which suggests a typical spin polaron character in the transport properties of $\text{La}_{1-x}\text{Ca}_x\text{MnO}_3$.

III. Properties of Composite Polaron at Finite Temperature

With the raise of temperature, more and more phonons and spin-waves are excited, whereas the increase of an external magnetic field decreases the spin-wave excitations. Thus the formation and the properties of the CP can be heavily affected by the external magnetic field \mathbf{B} and temperature T .

At finite temperatures, the thermal excitation of phonon and spin waves become

more and more important, their occupations obey the Bose-Einstein distribution law. We choose the following basis function for the CP at temperature T:

$$|\phi_i(S_0^z) \rangle = \sqrt{\frac{S + S_{0i}^z}{2S + 1}} [d_{i\uparrow}^\dagger + \frac{d_{i\downarrow}^\dagger S_i^\dagger}{S + S_{0i}^z}] |0 \rangle_e |n_i \rangle_{ph} |m_i \rangle_m \quad (10)$$

where $|0 \rangle_e$ denotes the electron vacuum state, $|n_i \rangle_{ph}$ the thermal-equilibrium phonon state and $|m_i \rangle_m$ the magnetic excitation state at site R_i at temperature T under magnetic field \mathbf{B} . Denoting a_i^\dagger (or a_i) as the creation (or the annihilation) operator of the CP, then the wavefunction of the CP can be expressed as:

$$|\Phi_i \rangle = \sum_i a_i |\phi_i(S_0^z) \rangle. \quad (11)$$

Expressing the Hamiltonian (5) in terms of the CP operators, we obtain

$$\tilde{H} = \sum_{i,j} a_j^\dagger a_i \langle \phi_j(S_0^z) | H' | \phi_i(S_0^z) \rangle \quad (12)$$

After a tedious calculation with the use of Fourier transformation, one gets:

$$\begin{aligned} \tilde{H} = & \sum_k [\epsilon_d - J_H S - \frac{2S}{2S+1} (\mu_B B + \frac{g}{2S} \mu_B B + A \sum_\delta S_\delta^z)] a_k^\dagger a_k \\ & + \sum_k z t e^{-p(\langle n_B \rangle + \frac{1}{2})} [1 + \frac{1}{2S+1} \sum_q (\frac{2S}{2S+1} \gamma_q - 1) \langle m_q \rangle] \gamma(k) a_k^\dagger a_k \end{aligned} \quad (13)$$

where $\langle n_B \rangle = 1/[\exp(\hbar\omega/k_B T) - 1]$ and $\langle m_q \rangle = 1/[\exp(\hbar\Omega_q/k_B T) - 1]$ denote the thermal-equilibrium mean occupations of the phonon and of the spin waves at temperature T, respectively; here $\hbar\Omega_q$ is the energy spectrum of the spin waves in double-exchange ferromagnets, $\hbar\Omega_q = g\mu_B B + 2zA(1 - \gamma_q)$. In the calculation, the linear approximation of the Holstein-Primakoff transformation:

$$S^\dagger = \sqrt{2S} c, \quad S^- = \sqrt{2S} c^\dagger, \quad S^z = S - c^\dagger c$$

was adopted. The average energies of the free phonons and of the free spin waves were not included.

Compare the dispersion of the CP in Eq. (13) with the bare energy spectrum $zt\gamma(k)$ of electrons, the bandwidth of the carriers at finite temperature T is renormalized by a factor of Z,

$$Z = e^{-p(\langle n_B \rangle + \frac{1}{2})} [1 - \frac{1}{2S+1} \sum_q (1 - \frac{2S}{2S+1} \gamma(q)) \langle m_q \rangle]. \quad (14)$$

One finds that the renormalized factor Z consists of two parts: the phonon renormalized part Z_{ph} and the spin-wave part Z_m , $Z=Z_{ph}Z_m$. Z exhibits strong temperature- and magnetic field-dependence. The phononic part,

$$Z_{ph} = \exp[-p(< n_B > + \frac{1}{2})] ,$$

depends only on the temperature. The increase of the electron-phonon interaction and temperature will narrow the conduction band further. The magnetic part,

$$Z_m = 1 - \frac{1}{2S+1} \sum_q [1 - \frac{2S}{2S+1} \gamma(q)] < m_q > ,$$

depends both on the temperature and the magnetic field. With the increase of temperature, the occupation of spin waves, $< m_q >$, becomes large. Since $(1 - \frac{2S}{2S+1} \gamma(q))$ is always positive, so the factor Z_m becomes small. Therefore in the region where temperature is below T_c , the bandwidth of the CP becomes narrower and narrower with the increase of temperature and the CP may be more easily trapped, so the mobility of the CP becomes smaller. Under strong magnetic field, the increase of field strength depresses the excitation of spin waves, reduces the number of spin waves surrounding the electron, thus the bandwidth of the CP becomes broader with external magnetic field and the mobility of the CP becomes larger. These properties coincide with experimental observations for resistivity. When T approaches the Curie temperature from below, the number of spin wave excitations reach its maximum and the factor Z_m approaches its minimum. When $T > T_c$, the above treatment of the linear spin wave approximation (via the Holstein-Primakoff transformation) is no longer true since the long-range ferromagnetic order does not exist anymore and one has to deal with the spin operators rather than the spin wave operators. Recent experimental observations [25] showed that the collective spin excitations also exist above the Curie temperature which suggested that the composite polaron can preserve for temperatures above T_c .

An interesting property of the CP is its transport properties in the presence of magnetic field B at finite temperature T , which is closely related to the transport properties of lanthanum manganite, especially the CMR effect. It is found that the magnetoresistance behavior of lanthanum manganites can be understood qualitatively

in the present theory of composite polaron. The electric resistivity can be expressed as

$$\rho = 1/(ne\mu), \quad \mu = e\tau/m^*$$

where μ is the mobility of the CP, m^* is the effective mass which is proportional to the inverse of the bandwidth. According to Eq.(13) and (14), one can determine the mobility through the effective mass, $\mu \propto 1/m^* \propto Z_m Z_{ph}$. At temperatures below the Curie temperature T_c , there exist both the spin-wave and the Jahn-Teller phonon excitations. As temperature arises, more and more spin waves and phonons are excited and the mobility of the CP becomes smaller. Consequently, the resistivity increases. The electric conductivity decreases with the increase of temperature and reaches its maximum near the Curie point. Above T_c , the long-range magnetic ordering disappears and only the Jahn-Teller phononic polaron plays a role. Like usual polarons, the hopping probability of the polaron to the nearby sites increases with the lift of temperature, so the resistance declines with the increase of T in high temperature region. Due to strong polaron self-trapping effect and its thermal activation, one would expect that a great change of the resistance with the increase of temperature, and that the depression of the magnetic field to the local spin fluctuation will reduce the bonding strength of the composite polaron, hence decrease the resistivity. From the preceding discussions, it is found that the narrowing of the electronic bandwidth is a fundamental process controlling the transport properties of conduction electrons in ferromagnetic narrow-band metallic manganites, a point supported by recent experiment [26].

One could estimate the magnetoresistance change through the mobility of the CP. The mobility variation comes from two parts, the effective mass and the scattering lifetime:

$$\frac{\mu}{\mu_0} = \frac{m}{m^*} \frac{\tau}{\tau_0} \quad (15)$$

here μ_0 and τ_0 denote the mobility and the scattering lifetime of free carrier. For typical electron-phonon coupling, $p \approx 1$, and the ferromagnetic coupling, $4zA = 30 \text{ meV}$ [20] when temperature ranges from 50 to 250 K, the variation of the contribution of the effective mass to the mobility of the composite polaron decreases about one order in magnitude, or, $m/m^* \sim 0.1$. The effect of magnetic field is not so significant as tem-

perature. At $T=250$ K, the the mobility contribution from the effective mass increases by two times when the magnetic field increases from zero to 8 Tesla. The reason is that the magnetic field only affects Z_m , however, temperature affects both Z_{ph} and Z_m . In fact, the huge change of the resistivity of the CP with magnetic field and temperature may come from the variation of the scattering lifetime, which can change several orders in magnitude and drive a metal-insulator-like transition in doped manganites. Detail studies is tedious and will be published elsewhere.

IV. Conclusion

To summarize, a theory of the composite polaron is developed. It is suggested that the composite polaron can be realized in lanthanum manganites, and contribute significantly to the unusual ground state, the thermodynamic, and the transport properties of manganites.

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